



Practitioner's Docket No.: 791\_130 RCE

**PATENT**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re the Application of: Li YANG and Toshihiro YOSHIDA

Serial No.: 09/770,725

Group Art Unit: 1746

Filed: January 26, 2001

Examiner: Jonathan Crepeau

Conf. No.: 6015

For: LITHIUM SECONDARY BATTERY

Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

"EXPRESS MAIL" mailing label number EV 48855 9447 US.

I hereby certify that this paper or fee is being deposited on  
APRIL 8, 2005 with the United States Postal Service  
"Express Mail Post Office to Addressee" service under 37 CFR §1.10  
addressed to: Mail Stop Amendment, Commissioner for Patents, P.O.  
Box 1450, Alexandria, VA 22313-1450.

*Janet M. Stevens*  
Janet M. Stevens

**DECLARATION UNDER 37 CFR §1.132**

Sir:

I, Toshihiro Yoshida, a citizen of Japan hereby declare and state:

1. I have a bachelor's degree in science which was conferred upon me by Tokyo University of Science in Tokyo, Japan, in 1988.
2. I have been employed by NGK INSULATORS, LTD. since 1988 and I have had a total of 7 years of work and research experience in the field of batteries, including lithium secondary batteries.
3. I am one of the inventors in the above-identified patent application and I am familiar with the references applied in the Office Action mailed January 14, 2005.

4. My publications include the following works in this field: "A model of capacity fade caused by a reaction rate between electrolyte and intercalated Li in carbon in Li-ion battery", Proceeding of The 43rd Battery Symposium in Japan (2002) and "A degradation mechanism and life prediction of Li-ion batteries", Proceeding of The 45th Battery Symposium in Japan (2004).

5. The following tests were conducted by me or under my direct supervision.

## REPORT OF EXPERIMENT

### Purpose of Experiment

The purpose of this experiment is to show the effect of HF in electrolyte solutions upon dissolution of transition-metal contained in lithium transition metal compound oxide, and to clarify the difference in the respective amounts of transition metal dissolved into electrolyte solution between lithium nickel oxide and lithium manganese oxide.

### Experimental

Experiment 1: amount of HF in electrolyte solution

Electrolyte solutions (20 ml) each consisting of 1 mol/L  $\text{LiPF}_6/\text{EC}+\text{DEC}$  (1:1 in volume) were heated up to 60°C or 100°C for 400 hours. The respective amounts of HF

in the electrolyte solutions thus heated were detected by a titration method, and are shown in Table 1.

#### Experiment 2: amount of transition metal dissolved

Powders (5g) of positive electrode material were soaked into the electrolyte solution (20 ml), the compositions of which are described above. Two types of the material were used: lithium nickel oxide ( $\text{LiNiO}_2$ ) and lithium manganese oxide ( $\text{LiMn}_2\text{O}_4$ ). The electrolyte solutions with powders were heated up to  $60^\circ\text{C}$  or  $100^\circ\text{C}$  for 400 hours. After filtration of the powders, the amount of transition metal in the electrolyte solution was detected by the ICP method, and are shown in Table 2.

#### Results and Discussion

Table 1: Amount of HF in Electrolyte Solution Heated

	Amount of HF (PPM)
$60^\circ\text{C}$ x 400 hours	313
$100^\circ\text{C}$ x 400 hours	2,474

Table 2: Amount of Transition Metal in Electrolyte Solution Heated

	$\text{LiNiO}_2$ Amount of Ni (PPM)	$\text{LiMn}_2\text{O}_4$ Amount of Mn (PPM)
$60^\circ\text{C}$ x 400 hours	3	22
$100^\circ\text{C}$ x 400 hours	4	5,600

Table 1 shows that HF is generated by heating of the electrolyte solution. This phenomenon is similar to that in which HF is generated by the addition of water into an electrolyte solution.

Table 2 shows that the transition metal constituting positive electrode materials dissolves into the electrolyte solution by heating. HF is considered to enhance the dissolution of transition metals. The dissolution rate of Mn is much higher than that of Ni. The difference indicates that the amount of dissolution of transition metal into the electrolyte solution strongly depends on the respective type of positive electrode material.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon

Date: 29 March, 2005

By: T. Yrshater